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Measurements of SO₂ profiles in volcanic plumes from the NASA Tropospheric Emission Spectrometer (TES)

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[1] Satellite measurements are now recognized as a key element for the early detection and characterization of volcanic eruptions, in particular in the context of aircraft routing. A common tracer of volcanic plumes is sulfur dioxide (SO₂), which so far has been measured by ultraviolet-visible (UV-vis) instruments and multispectral infrared (IR) sounders. Here we report the first SO₂ vertical profile retrieved from high spectral resolution thermal infrared nadir radiance spectra and we provide information on both the quantity of gas emitted and its altitude. From the radiance spectra provided by the Tropospheric Emission Spectrometer (TES) aboard the NASA AURA satellite, and owing to the $\sim 0.1 \text{ cm}^{-1}$ (apodized) spectral resolution, elevated levels of SO₂ were measured following volcanic eruptions occurring in 2005 (Manam, Sierra Negra) and 2006 (Rabaul, Nyamuragira). Column values are found to be in good agreement with the data provided by Ozone Mapping Instrument (OMI), a UV-vis instrument also onboard the AURA satellite. **Citation:** Clerbaux, C., P.-F. Coheur, L. Clarisse, J. Hadji-Lazaro, D. Hurtmans, S. Turquety, K. Bowman, H. Worden, and S. A. Carn (2008), Measurements of SO₂ profiles in volcanic plumes from the NASA Tropospheric Emission Spectrometer (TES), *Geophys. Res. Lett.*, 35, L22807, doi:10.1029/2008GL035566.

1. Introduction

[2] Emissions of gases and particles associated with intense volcanic eruptions can ascend to the upper troposphere and stratosphere where they can alter atmospheric chemistry and climate [Robock, 2000]. Tracking the evolution of volcanic plumes is crucial to aviation hazard mitigation because ash and sulfuric acid droplets can diminish visibility, damage flight control systems, and cause jet engines to fail. Key observations are provided by satellite-borne instruments as they can routinely monitor the downwind transport of gases and aerosols following an eruption, and the spatial extent of the volcanic cloud. Common tracers of volcanic plumes are ashes and sulfur

dioxide (SO₂), which can be tracked up to several days after the eruption. The plume can split following different trajectories, with a higher SO₂ rich and a lower ash rich part [Prata and Kerkmann, 2007].

[3] Satellite instruments operating in the ultraviolet (UV) spectral region (e.g. TOMS, GOME, OMI) [Krueger *et al.*, 1995; Khokhar *et al.*, 2005; Carn *et al.*, 2007] have good sensitivity to SO₂ but only deliver total column amounts during daytime. Infrared (IR) multispectral imaging sounders (MODIS, AVHRR, TOVS, ASTER) provide an alternative to monitor volcanic plumes from space [Prata, 1989; Prata *et al.*, 2003; Pugnaghi *et al.*, 2006; Watson *et al.*, 2004]. However, their broadband spectral channels prevent accurate determination of the temperature distribution and do not allow a proper discrimination of the SO₂ signal from strong interfering water vapor and aerosol signatures [Realmutto and Worden, 2000]. Recently, Carn *et al.* [2005] and Eckhardt *et al.* [2008] have shown, using AIRS/AQUA satellite data, that advanced meteorological thermal IR sounders could significantly contribute to the tracking of volcanic SO₂ plumes. In this work, we further investigate how much information is added to the analysis of volcanic plumes using the higher spectral resolution measurements provided by the Tropospheric Emission Spectrometer (TES) [Beer, 2006]. We estimate the SO₂ vertical profile, which is good indication of the plume's altitude and therefore provides critical information for aircraft routing. Our analysis is performed for a set of four representative volcanic events in 2005 (Manam, Papua New Guinea [PNG]; Sierra Negra, Galapagos Islands) and 2006 (Rabaul, PNG; Nyamuragira, DR Congo).

2. TES Instrument Onboard AURA

[4] TES is one of four instruments onboard NASA's AURA satellite launched in July 2004. TES is a Fourier Transform Spectrometer (FTS) designed to infer the vertical distribution of tropospheric ozone and carbon monoxide using both nadir and limb viewing geometries from outgoing spectrally resolved IR radiation. During routine operation, the radiometrically calibrated nadir spectra [H. Worden *et al.*, 2006] cover the thermal IR region in four bands from 652–919 (band 2B1), 923–1160 (1B2), 1090–1339 (2A1) and 1891–2251 (1A1) cm^{-1} at a spectral resolution of 0.1 cm^{-1} (apodized), which are shown in Figure 1 (inset). The AURA spacecraft is in a sun-synchronous orbit at an altitude of about 705 km, with a 13:38 local mean solar time ascending node. For the nadir observation, TES employs 1×16 linear arrays of rectangular pixels (each is $5 \times .5 \text{ km}$). The projections of these arrays used in nadir are 8 km along-track and 5 km cross-track at the Earth's surface.

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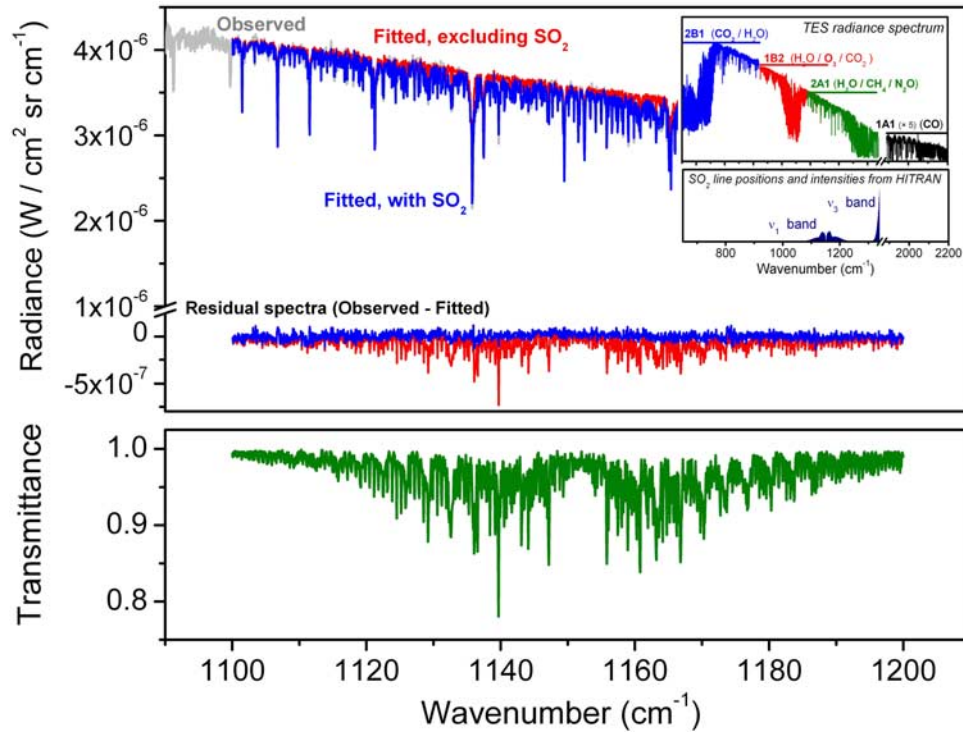


Figure 1. (top) TES radiance spectrum (average of 16 individual pixels, in $\text{W}/\text{cm}^2 \text{sr cm}^{-1}$) in the vicinity of Nyamuragira volcano (0.13°N – 25.64°E), on November 28, 2006. The grey line is the measured atmospheric spectrum. The blue and red lines are the adjusted spectra and residuals with and without the SO₂ contribution taken into account, respectively. (bottom) SO₂ contribution to the measured spectrum, in transmittance units. The inset shows on the top a radiance spectrum measured by TES, with the different spectral bands and dominant absorbing species labeled, and on the bottom the line integrated absorption cross sections (cm molecules^{-1}) for SO₂ in the ν_1 and ν_3 vibrational bands, from the HITRAN 2004 database.

[5] The vertical distribution of the following atmospheric products are retrieved operationally: atmospheric temperature and H₂O [Bowman *et al.*, 2006], CO [Rinsland *et al.*, 2006], O₃ [Bowman *et al.*, 2006; Worden *et al.*, 2007], and HDO [J. Worden *et al.*, 2006]. This work investigates the possibility of retrieving SO₂ from TES data for the first time.

3. Results: SO₂ Profiles and Total Columns During Recent Volcanic Eruptions

3.1. Methodology for SO₂ Profile Retrievals

[6] SO₂ absorbs thermal IR radiation in the ν_1 band around 1150 cm^{-1} , the ν_3 band around 1350 cm^{-1} , and the $\nu_1 + \nu_3$ band around 2500 cm^{-1} . The ν_3 band is the most prominent but lies in a range where strong absorptions by methane and water vapor occur, which makes it difficult to accurately retrieve tropospheric SO₂ concentrations [Carn *et al.*, 2005; Realmuto and Worden, 2000]. It is furthermore only partly covered by TES on the longwave end of band 2A1 (see inset of Figure 1). In this work, we therefore use the radiance signal in the ν_1 band to retrieve SO₂ profiles.

[7] The SO₂ profile and total column retrievals are performed using the *Atmosphit* software, developed at ULB [Coheur *et al.*, 2005; Clerbaux *et al.*, 2005], which contains ray tracing for various geometries, a line-by-line

radiative transfer model, and an inversion scheme that relies on optimal estimation [Rodgers, 2000]. A synthetic spectrum is computed using the line parameters and absorption cross sections for the heavier molecules, as collected in the HITRAN 2004 database [Rothman *et al.*, 2005]. The resulting spectrum is then processed to account for the TES Instrumental Line Shape [Clough *et al.*, 2006].

[8] The *Atmosphit* software provides the derivatives of the radiances \mathbf{y} with respect to the vertical abundances of the SO₂ target species: the Jacobians $\mathbf{K} = \partial \mathbf{y} / \partial \mathbf{x}$. Starting from relevant *a priori* information, composed of a mean state \mathbf{x}_a , and an *a priori* covariance matrix, \mathbf{S}_a , which represents the best statistical knowledge of the state prior to the measurements, the retrieved state can then be found. We used a standard SO₂ *a priori* profile from AFGL 1976 and the \mathbf{S}_a matrix was set proportional to the identity matrix.

[9] Assuming a linear problem, the optimal vertical profile $\hat{\mathbf{x}}$ can be written as [Rodgers, 2000]:

$$\hat{\mathbf{x}} = (\mathbf{K}^T \mathbf{S}_\epsilon^{-1} \mathbf{K} + \mathbf{S}_a^{-1})^{-1} (\mathbf{K}^T \mathbf{S}_\epsilon^{-1} \mathbf{y} + \mathbf{S}_a^{-1} \mathbf{x}_a) \quad (1)$$

where \mathbf{S}_ϵ is the measurement noise covariance matrix. In this work, \mathbf{S}_ϵ is diagonal and chosen in a conservative manner, as the maximum value of the signal to noise in the

Table 1. SO₂ Total Column and Retrieved Altitude of Peak Concentration as Measured by TES and OMI Onboard AURA for Several Eruptive Volcanic Events in 2005 and 2006^a

Volcano, Date	Coordinates	Total SO ₂ Column From TES (DU)	Retrieved Peak From TES (km)	Total SO ₂ Column From OMI (DU)	Altitude Plume (est.) (km)
Manam 28.01.2005	3.39°S–142.17°E	25	16	23(STL)	21–24
Sierra Negra 24.10.2005	4.89°S–97.05°W	16	3	1.4(PBL)/0.10(TRL)	5
	3.31°S–97.38°W	31	3	9.8 (PBL)/2.78(TRL)	
	1.59°S–97.75°W	46	3	68.1(PBL)/30.8(TRL)	
Rabaul 08.10.2006	19.67°S–161.09°E	6	16	5.7 (STL)	18
	3.26°S–157.29°E	2	16	4.1 (STL)	
	1.54°S–157.12°E	3	15	4.7 (STL)	
	0.10°N–156.77°E	3	15	4.4 (STL)	
	1.68°N–156.43°E	2	16	2.6 (STL)	
Nyamuragira 28.11.2006	0.13°N–25.64°E	110 (75–164)	2.5 & 15	298(PBL)/128(TRL)	3 & 15
	1.65°N–22.21°E	42 (11–62)	2.5 & 15	7.42(PBL)/1.78(TRL)	

^aSO₂ given in Dobson units. Dates given in dd/mm/yyyy. In the case of the intense Nyamuragira eruption, where SO₂ was retrieved from the individual TES pixels, both the mean and the range of retrieved columns are reported. At each location, the collection 3 AURA-OMI Level 2 sulfur dioxide product is available for 4 a-priori assumed SO₂ profiles: STL (lower stratosphere; SO₂ located between 15 and 20 km), TRM (mid-troposphere; SO₂ located between 5 and 10 km), TRL (lower troposphere; SO₂ located between 0 and 5 km) and PBL (SO₂ located in the planetary boundary layer). The last column gives the estimated plume altitudes as reported in *Carn et al.* [2008] and *Yang et al.* [2007].

TES 2A1 spectral band. Using this formalism, we can also derive the averaging kernel matrix, defined as

$$\mathbf{A} = \frac{\partial \hat{\mathbf{x}}}{\partial \mathbf{x}} = \mathbf{GK}$$

The vertical resolution of the retrieved profile can be defined as the Full Width at Half Maximum (FWHM) of the rows of the averaging kernel matrix. The number of statistically independent elements of information contained in the measurement can also be estimated as the Degrees Of Freedom for Signal (DOFS) which is defined as the trace of the averaging kernel matrix [Rodgers, 2000]. The total error is computed as the difference between the true state and the retrieved state. The smoothing error (smoothing of the true state by the averaging kernels) is found to be the dominant error here. The pressure, temperature and H₂O profiles, as well as information on cloud top and average optical depth, were taken from the TES Level 2 operational processing. The concentration of all interfering species (H₂O and to lesser extent CH₄, N₂O) was also retrieved to allow the best achievable fit in the spectral band of interest.

3.2. SO₂ From TES and Comparison With OMI

[10] A systematic search for intense SO₂ absorption signatures in the TES spectra was undertaken, in particular in the vicinity of strong volcanic events that occurred in 2005 and 2006. In order to select the radiance data from which SO₂ might tentatively be retrieved, we relied on observations reported by other satellite missions (ASTER, OMI). To illustrate, Figure 1 shows a TES radiance spectrum recorded over Nyamuragira volcano, on November 28, 2006. The spectral absorption signature of the SO₂ ν_1 band centered around 1150 cm⁻¹ can clearly be seen from the residual plot obtained with and without taking the SO₂ contribution into account in the radiative transfer simulation.

[11] As the horizontal coverage of TES is limited, some major volcanic events measured by other satellite sensors might be missed when the instrument's narrow nadir swath

does not intersect the volcanic plume. Four eruptions occurring in 2005 and 2006 (Manam and Sierra Negra in 2005; and Rabaul and Nyamuragira in 2006) were found to be observable in TES radiance spectra. We analyzed the TES spectra recorded in the area of the volcano and estimated SO₂ vertical profiles and derived column amounts for each case. From the four events analyzed in this work, an upper bound for the detection of SO₂ in ν_1 from TES is 2 Dobson Units (DU). These measurements are reported in Table 1, along with the location and time of each observation. In general we used the average of the 16 pixels as recommended by the TES science team, in order to increase the signal to noise ratio, but the absorption signal found in the Nyamuragira eruption cloud was so intense that for this event we were able to work on the individual pixels, and the results are presented in terms of a range of total column values and the associated mean. Table 1 also lists the corresponding OMI total column SO₂ retrievals, as well as the assumed plume height used for the OMI retrievals. Figure 2 provides a color representation of the SO₂ total column amounts as measured from TES and OMI, in the vicinity of the Rabaul volcano.

[12] Overall, we find a very good correspondence between OMI and TES retrieved total columns for the Manam and Rabaul eruptions, characterized by high altitude plumes that reached the lower stratosphere. Discrepancies for low altitude plumes can come in part from the much larger pixel size of OMI, which tends to dilute the SO₂ signal, but also from the less accurate TES estimation of SO₂ amounts in the lowest layers of the atmosphere. For the retrievals from Nyamuragira, for which the peak in the SO₂ profile occur at both 2.5 and 15 km, the comparison is difficult, as OMI shows total column amounts strongly dependent on the assumed plume altitude. More on this can be seen in Figure 3, which compares retrieved absorbance spectra as well as associated vertical profiles from the Manam and Sierra Negra eruptions. Although the retrieved total column for Manam is about half that of Sierra Negra, the SO₂ absorbance is significantly larger in the Manam case due to the higher altitude of the plume, which produces narrower

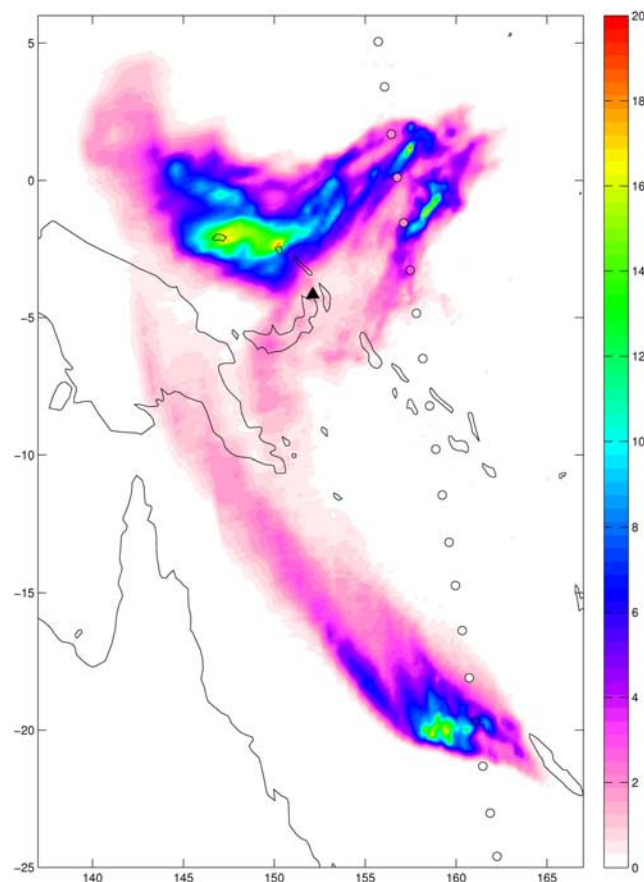


Figure 2. Total column measurements of SO₂ (DU) on 8 October for the Rabaul eruption, as measured by OMI (colored clouds) and TES (circles) onboard AURA in 2006. The location of the volcano is indicated by a black triangle.

and stronger lines (Figure 3, top). It is this effect, combined with the well documented loss of sensitivity of thermal IR instruments to the boundary layer, that results in large errors on the retrieved SO₂ concentrations for low altitude plumes (Figure 3, bottom).

[13] As is apparent from Figure 3, the high spectral resolution of TES makes it possible to discriminate between low and high altitude SO₂ plumes on the basis of the line widths, which increase with increasing (decreasing) pressure (altitude). Depending on the plume, we found that the TES measurements contain information on the SO₂ vertical profile with a few (4 to 6) kilometers vertical resolution. We measured the altitude of Rabaul and Manam plumes to be around 16 km, which is compatible with the estimate provided by *Carn et al.* [2008] of 18 km for Rabaul and 21–24 km for Manam. Similarly for Nyamuragira, for which our retrieved profile shows a double peak structure with maxima at 2.5 and 15 km, and Sierra Negra, for which our estimated plume altitude is at 3 km, we are in the range of the values reported by *Carn et al.* [2008] and *Yang et al.* [2007], i.e. altitudes between 3 and 15 km for Nyamuragira and 5 km for Sierra Negra.

4. Conclusion and Perspectives

[14] In this paper we have presented the first detailed retrievals of SO₂ using the thermal IR high spectral resolu-

tion TES/AURA measurements, for four different volcanic plumes. The UV-visible OMI sensor onboard the same platform provides routine measurements of SO₂ (<http://so2.umbc.edu/omi/>) with better spatial coverage, but is limited to one daylight observation per day (except at high latitudes in summer), and requires an estimate of the SO₂ plume altitude for accurate retrievals. TES measurements, although limited in spatial coverage, can add valuable information to OMI measurements to derive the SO₂ atmospheric loading, as it provides additional nightly measurements, as well as plume altitude estimation.

[15] With respect to total columns we found a good agreement with OMI for the high altitude plumes (around 25% on average). Larger differences were found for some of the lower altitude plumes, possibly due the larger pixel size of OMI and the inherent lower sensitivity of thermal IR sounders near the surface.

[16] The high spectral resolution of TES allowed us to derive vertical profiles of SO₂. Peak altitudes were found to be in good agreement with the literature. From a spectroscopic point of view, this is the first time vertical profiles have been reported using the weak ν_1 band of SO₂, which is made possible by the high spectral resolution of TES.

[17] The tools developed in the framework of this study will be applicable to AIRS and IASI [*Clarisse et al.*, 2008], a new thermal infrared FTS carried onboard the METOP satellite. Despite their coarser spectral resolution as com-

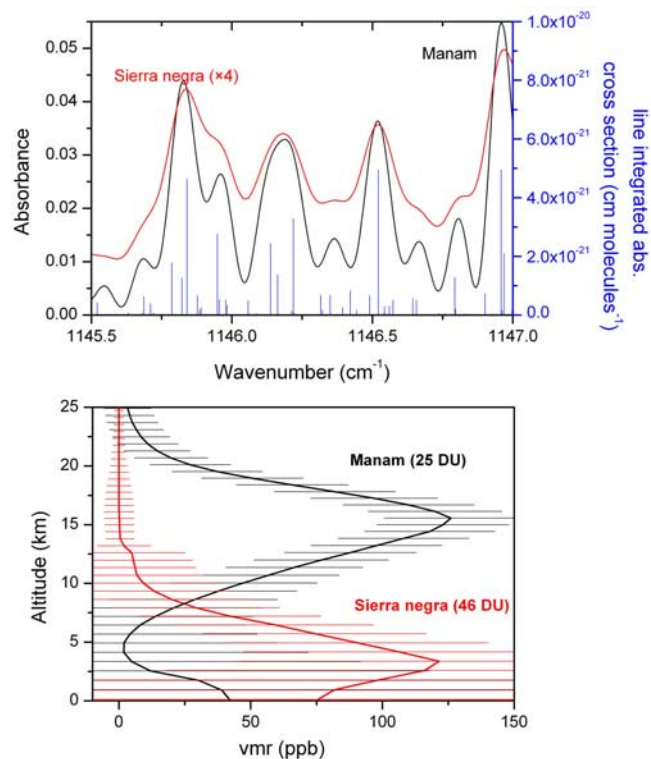


Figure 3. (top) Line integrated absorption cross sections (cm molecules⁻¹) for SO₂ (blue) and absorbance spectra from the Sierra Negra (red) and Manam plume (black). These absorbance spectra were calculated as $A = -\ln(\tau)$, where τ is the retrieved transmittance due to SO₂. (bottom) Retrieved profiles from Sierra Negra (red) and Manam (black) and associated indicative retrieval error bars.

pared to TES, these thermal infrared sounders have the advantage to be on meteorological platforms, thereby offering global coverage twice daily with a relatively small ground pixel size. They are for these reasons more adapted than TES to provide operational information on volcanic activity and to follow the evolution of the plumes in space and time. Current and planned applications of AIRS and IASI include aviation hazard mitigation.

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